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Techno-economic analysis of ammonia production via integrated biomass gasification

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HIGHLIGHTS

- Techno-economic results regarding biomass-based ammonia production systems.
- Integration of an ammonia production process in a pulp and paper mill.
- Integrated ammonia production gains higher system efficiency than stand-alone production.
- The economics of an integrated production system is improved compared to stand-alone production.

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ABSTRACT

Ammonia (NH_3) can be produced by synthesis of nitrogen and hydrogen in the Haber–Bosch process, where the economic challenge is the hydrogen production. Currently, substantial amounts of greenhouse gases are emitted from the ammonia industry since the hydrogen production is almost exclusively based on fossil feedstocks. Hydrogen produced via gasification of lignocellulosic biomass is a more environmentally friendly alternative, but the economic performance is critical. The main objective of this work was to perform a techno-economic evaluation of ammonia production via integrated biomass gasification in an existing pulp and paper mill. The results were compared with a stand-alone production case to find potential technical and economic benefits deriving from the integration. The biomass gasifier and the subsequent NH_3 production were modelled using the commercial software Aspen Plus. A process integration model based on Mixed Integer Linear Programming (MILP) was used to analyze the effects on the overall energy system of the pulp mill. Important modelling constraints were to maintain the pulp production and the steam balance of the mill. The results showed that the process economics and energy performance are favourable for the integrated case compared to stand-alone production. The main conclusion was however that a rather high NH_3 selling price is required to make both production cases economically feasible.

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1. Introduction

The fertilizer sector annually stands for 1.2% of the world's total energy use, where the bulk (87%) is directly coupled to ammonia (NH_3) production [1]. NH_3 is one of the most produced inorganic chemicals worldwide, with a total global production of 109 Mt in 2009 [2] and the global production capacity is forecasted to increase to nearly 239 Mt by 2020 [1]. NH_3 can be produced via synthesis of nitrogen and hydrogen in the Haber–Bosch process, where the economic challenge is the hydrogen production. Currently, substantial amounts of greenhouse gases are emitted from the NH_3 industry since the hydrogen production is almost exclusively based on fossil feedstocks. The main hydrogen source was previously gasification of coal, but falling petroleum prices shifted the production to steam reforming of natural gas [3]. A more environmentally friendly option would be to produce the hydrogen via thermal conversion of lignocellulosic biomass.

Existing ammonia plants usually have very large production capacities, typically exceeding 1500 tons of ammonia per day. To be competitive, it is likely that similar production capacities will be required for biomass based ammonia plants. Such a production level would approximately require in the order of 2000 tons of dry biomass per day, which puts high demands on biomass logistics and handling. The production capacity in practice will however be constrained by the available biomass supply and gasifier scales [2].

Gilbert et al. [2] have made a review of different biomass gasifiers in order to find the best suited technology for ammonia production. In that study, a Fast Internal Circulating Fluidised Bed gasifier

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Nomenclature

AGR	acid gas removal	LP	low pressure
ASU	air separation unit	MILP	Mixed Integer Linear Programming
BAU	business-as-usual	MP	medium pressure
BB	bark boiler	PEBG	pressurized entrained flow gasifier
BIGCC	biomass integrated gasification combined cycle	PI	process integration
FICFB	Fast Internal Circulating Fluidised Bed	RB	recovery boiler
HP	high pressure	S-A	stand-alone
IRR	internal rate of return	WACC	weighted average cost of capital

(FICFB) with CO_2 removal was selected due to its generation of a hydrogen rich syngas. This gasification technology is currently demonstrated in the scale of 8-10 MW [3,4]. Up-scaling to a production capacity exceeding 100 MW will be a great challenge and probably not economically feasible without transition to a pressurized concept. One issue is that the FICFB technology is more difficult to pressurize than direct gasifiers [5]. Therefore, a pressurized entrained flow biomass gasifier (PEBG) has been chosen in this study as this technology also produces a high quality and hydrogen rich syngas, but has a much better scale-up potential than FICFB gasifiers.

Integrating biomass gasification processes for large-scale chemical production in existing industries may result in several technical, energy-related as well as economic benefits, e.g. [6–8]. A large number of studies regarding integrated chemical production in pulp and paper mills have recently been published [9–12]. One reason why this industry is of such a great interest is the already existing biomass supply infrastructure, which provides large logistical and feedstock handling advantages. Furthermore, the industry is facing increasing energy prices and stronger competition on raw materials [13], which makes it important to find additional streams of revenues to their existing production.

The main aim of this work was therefore to perform a techno-economic evaluation of NH_3 production via an integrated PEBG in an existing pulp and paper mill. The main objectives were to:

- Investigate if this production concept is economically feasible at current market conditions.
- Find measurable technical and economic added values for integrated biomass based ammonia production compared to stand-alone units.

2. Material and method

A process integration (PI) model was used to evaluate the integration of gasification based NH_3 production in an existing pulp and paper mill. The most important modelling constraint was to maintain the pulp production and consequently the process steam balance. A detailed Aspen Plus process model was used to model energy and material balances of the complete biomass gasification system including the NH_3 synthesis. An iterative modelling approach between the two models was adopted to ensure that all constraints of the pulp and paper mill as well as for the gasification plant were met. The resulting material and energy balances were used to analyze the different system configurations in terms of overall energy efficiency and process economics.

2.1. The process integration model

The process integration model was based on mathematical programming based on Mixed Integer Linear Programming (MILP). The model is composed of branches and nodes, where each node represents process units and energy/material flows, respectively. The different nodes are connected depending on the input and output to/from each process unit. Each node contains linear equations to express the energy and mass balance required in the process unit. Thus an entire energy system is created. The model of the existing mill configuration was validated against operational data. The model development and validation are further described in [14].

2.2. Modelling of the biomass gasification system

The gasifier model was based on the PEBG technology and created in Aspen Plus. Entrained flow gasifiers usually operate in a temperature range of 1200–1800 °C and at pressures in range of 20–70 bar [15]. These operating conditions yields a nearly tar and methane free syngas. The fuel was fed into a heated reactor co-currently with a gasifying agent, in this case pure oxygen (O_2), for partial combustion of the fuel [15,16].

The O_2 was produced via a cryogenic air separation unit (ASU) with an estimated power demand of 1.0 MW_e per kg-O_2/s (assuming a 99.6 mol% O_2 purity) [17].

The short residence time for the entrained fuel in the reactor requires particle sizes smaller than 0.5 mm, in order to facilitate high carbon conversion rates [16]. The water content and the particle size of the fuel must therefore be reduced before it is supplied to the reactor.

The fuel drying and the grinding were modelled as black-boxes. The biomass was specified according to the ultimate and proximate analysis of the fuel. The dryer was modelled as an indirectly heated rotary dryer, with a specific power consumption of 15 kWh per wet ton of biomass [18] and the water content of the biomass was assumed to be reduced from 40% to 8%. According to [19], in between 2% and 3% of the thermal input of the biomass (based on LHV) is estimated to be required in terms of specific electricity consumption for the grinding to 0.5 mm particle diameters. In this study, 3% was assumed. The biomass powder was assumed to be pressurized using screw feeders, which require an inert gas as well as electricity. The specific electricity consumption was estimated to 0.02 kWe per kW_{th} of biomass for pressurization up to 40 bar [20].

Nitrogen (N_2) is usually an undesired inert gas in syngas applications, but not during NH_3 synthesis, as it is produced by the reaction between N_2 and H_2 [21]. A hydrogen to nitrogen molar ratio of 3:1 is optimal for NH_3 synthesis [22], but this ratio could not be controlled if air were used as a gasifying agent.

The gasification process was modelled in detail by treating the dominant reaction steps in the gasifier separately, i.e. drying, pyrolysis, combustion and gasification. The pyrolysis step was modelled using empirical relationships by an approach developed and described in [23]. Furthermore, equilibrium restrictions were implemented to prevent underestimations of the methane and tar contents of the outgoing syngas. This approach provides a more accurate simulation of a pressurized entrained flow reactor [24]. A gasification pressure level of 30 bar (a) was assumed in the model. The temperature of the raw syngas leaving the reactor before entering the quench was assumed to be 1200 °C. In the quench, the gas was cooled to 500 °C after passing water jets and bubbling through a water reservoir. After the quench, the residual heat was recovered for steam generation.

2.3. Modelling of the gas cleaning, conditioning and ammonia synthesis

The optimal H_2:N_2 molar ratio is 3 for NH_3 synthesis [22] and to increase the hydrogen concentration of the syngas, a water gas shift (WGS) reactor was used. An acid gas removal (AGR) unit was placed after the shift reactor in order to remove any acid gas contaminations and CO_2. The AGR unit used cooled methanol as a physical solvent to remove unwanted impurities and gas species and is similar to the Rectisol® process [17].

As a final purification step, a liquid N_2 wash was used. The N_2 wash was located downstream the AGR unit and use the already cooled gas stream (below −50 °C). Methanol and the remaining traces of CO_2 in the syngas must be removed before the N_2 wash to avoid freezing problems in the cold box heat exchanger [22]. The main task for the N_2 wash is to remove the remaining CO content, because it acts as a poison for the NH_3 catalyst [22]. Pressurized N_2 and syngas were cooled against the product streams (outgoing syngas and off-gases). As a result, the N_2 was liquefied and then fed to the top of the wash column. The cooled syngas entered at the bottom of the column, where inert gases such as CH_4 and argon will condensate from the syngas whilst the liquefied N_2 removes the CO. An additional nitrogen stream was mixed with the outgoing syngas to reach the desired H_2:N_2 ratio of 3 [22].

The syngas was pressurized to the operating pressure of the NH_3 catalyst (180 bar) before entering the synthesis loop. The NH_3 synthesis took place over an iron promoted catalyst [21], modelled using a Gibbs reactor with an outgoing gas temperature of 440 °C. Typically the conversion efficiency is of the order of 20–30% per pass, and recycling of the unreacted gases is therefore necessary to increase the NH_3 production yield [25]. The NH_3 reaction is exothermic, meaning that cooling of the outgoing gases can generate high quality steam [21]. The produced NH_3 is separated from the unreacted gases by condensation in a refrigeration system. Part of the recycled gas must be purged to avoid inter gas accumulation in the synthesis loop [21]. To compensate for the pressure drop over the catalyst, the recycled gas was recompressed to 180 bar before it mixing with the incoming syngas.

A simplified description of the complete ammonia production process is shown in Fig. 1.

3. Case descriptions

The Billerud Karlsborg AB pulp mill, located in the northern part of Sweden, was used as case basis. The mill consists of two major processing lines: the fibre line and the chemicals recovery loop.

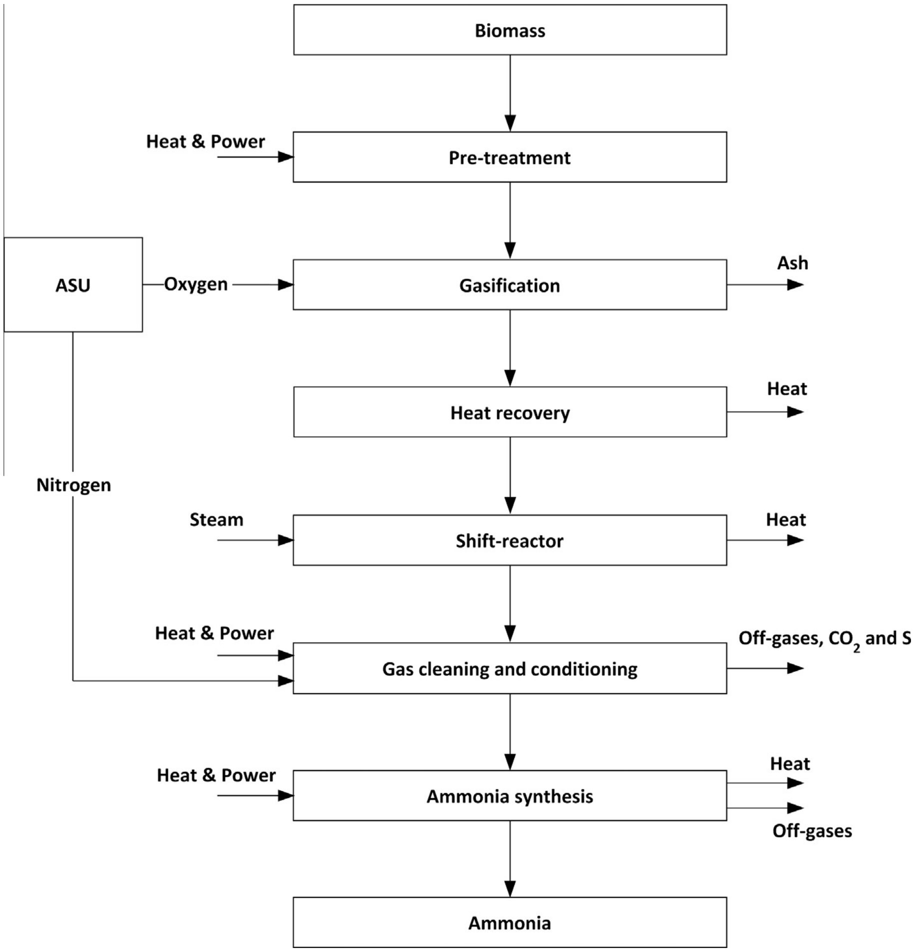

Fig. 1. Schematic model of the gasification based ammonia production plant.

The fibre processing line extends from the digester to the outgoing pulp and paper, see Fig. 2.

In the processing line, cooking chemicals (white liquor) and heat are used to dissolve the binding agent in wood (lignin) in order to release the cellulose fibres needed as raw material for the pulp and paper. The black liquor (BL) is a by-product from pulp making and contains spent cooking chemicals and organic matter. The recovery of these chemicals is necessary to make the process economically feasible and to create a closed chemical recovery cycle. The solid content of the BL is concentrated in multi-effect evaporators before being burned in a recovery boiler (RB). The combustion of organics provides energy to produce high pressure (HP) steam and to carry out the reduction reactions to recover cooking chemicals. Sodium hydroxide (NaOH) and sodium sulphide (Na_2S) are the main component in the white liquor solution, generated by passing the recovered cooking chemicals through the causticizing plant. In process of concentrating the BL, rosin soap is precipitated. The rosin soap is further processed into tall oil and used as fuel in the lime kiln. More HP steam is produced in the bark boiler (BB) using the internal surplus of the bark with the required extra biomass (or oil) to cover the steam demand of the mill. The produced HP steam expands to medium pressure (MP) and low pressure (LP) steam in a back-pressure turbine. A small quantity of the steam is extracted at 30 bar for soot-blowing of the RB, while MP and LP steam are consumed in the internal processes in the pulp and paper mill. The back-pressure turbine has a power output of 35 MW_e.

The original operation of the mill in combination with a stand-alone gasification based NH_3 production unit was served as a reference case (BAU/S-A). The surplus heat from the NH_3 plant was assumed to be sold to a local district heating network.

The alternative system configuration, titled Case A, was to replace the bark boiler in the pulp mill in favour of a PEBG unit for NH_3 production. As previously mentioned, the most important constraint was to maintain the pulp production. The integration was therefore done with the constraint that the MP and LP steam balance of the mill was kept. The bark originally used as fuel in the bark boiler was used as fuel in the gasification plant. Furthermore, it was assumed that off-gases from the synthesis loop together with tail gas from the N_2-wash could substitute the tall oil used in the lime kiln. The surplus tall oil can be sold on the market as an alternative to heavy fuel oil.

The stand-alone gasification plant has the same thermal gasification capacity and NH_3 production as in Case A.

3.1. Overall energy system analysis

The energy system efficiency was calculated using electricity equivalents. All energy carriers were converted to their corresponding electricity equivalents according to the efficiency of the best-available technologies known to the authors, see Table 1. The material and energy balance for Case A was calculated on an incremental basis compared to the operation of the pulp mill prior to the integration, i.e., required marginal supply of biomass and other energy carriers needed to produce NH_3. Case A was compared to the original operation of the mill and the stand-alone gasification plant (BAU/S-A) to analyze the effects on the overall energy system and to determine the best alternative for producing NH_3 from biomass gasification (i.e. non-integrated or integrated).

The biomass dedicated for pulp making and the final pulp products were not accounted for when the overall energy system efficiency were calculated as that supply is equal in both cases. The internal by-product bark becomes available from the debarking process of the pulp wood. Bark is an unwanted biomass sort in the cooking process due to its high ash content and other contaminations. The bark was therefore not accounted for as pulp making biomass. The system boundaries used for the overall system efficiency calculations is illustrated by the dashed boxes in Fig. 3.

3.2. Economic calculations and sensitivity analysis

In the economic calculations, the incremental investment, calculated as the difference between the investment in a new bark boiler and the investment in an integrated gasification plant for NH_3 production, was used in Case A. This implies that the bark

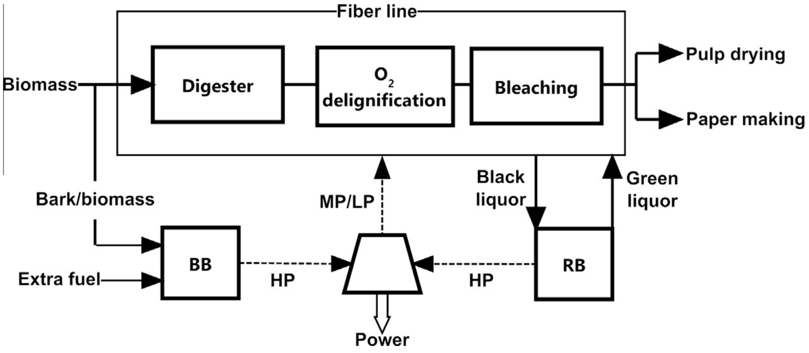

Fig. 2. Conventional fibre line and chemical looping.

Table 1
Power generation efficiencies used for calculation of electricity equivalents.

Energy carrier	Power generation efficiency (%)	Comment	Refs.
Biomass	46.2	BIGCC	[9]
Methanol	55.9	Gas turbine combined cycle	[9]
Tall oil	55.9	Assumed the same as methanol	
Hot water, 95 °C	10	Opcon power box	[9]
NH_3	49	Converted by multiplying the methanol power generation efficiency with the ratio of the lower heating values between ammonia and methanol	

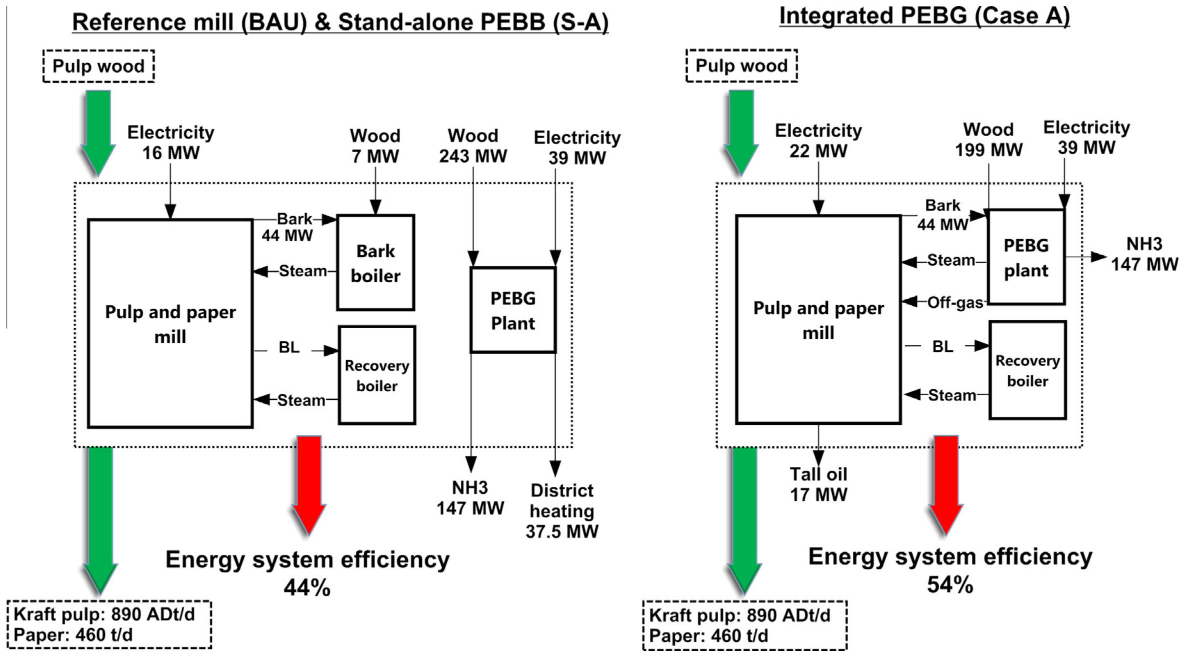

Fig. 3. Resulting energy and material balances. The system boundary includes the P&P plant and the stand-alone gasification unit (left side) and the complete integrated biomass gasification plant (right side). Biomass for pulp making and the final pulp products (green arrows) are the same for both cases and are neither accounted for in the efficiency calculation (air dry tons = AD/t). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

boiler of the mill is old and needs to be replaced in any case. The energy balances for Case A include the operational changes in the paper and pulp mill caused by the integration of the gasification plant. Cost and revenues associated with the original operation of bark boiler was included in the reference case, where the cost of operation and maintenance was taken from the "Mill stand-alone case" presented in [12].

Natural gas is the currently the main feedstock for NH_3 production [22], meaning that the price of natural gas is a determining factor of the NH_3 market price. Other factors, presumably the biomass price, would control the NH_3 market price if the production was primarily based on biomass. Therefore, an estimation of the required NH_3 selling price that would make biomass based NH_3 production an attractive investment opportunity was made. The NH_3 selling price was calculated via cash flow analysis at three levels of internal rate of returns (IRR). These required selling prices were also compared to the present average market selling price for fossil NH_3 (514 €/ton) [26]. In the cash flow analysis, a plant start-up period was considered, where the NH_3 production was 25% of the maximum capacity during year one and thereafter increased by 25%-points per year until the maximum production capacity was reached. The investment cost was assumed to be paid during a three year construction period. Costs and revenues from other products (electricity, tall oil, district heating, etc.) were included in the production cost of NH_3. The required NH_3 selling price to receive an internal rate of return (IRR) of 10%, 15% or 20% was investigated. In the economic evaluation, the NH_3 export was assumed to be exempted from taxes. Biomass for pulp making and the final pulp products were, as previously mentioned, not included in economic evaluation.

The investment cost for equipment and assembly was calculated as the over-night installation cost by the sum of the major units (pre-treatment, gasifier, AGR, etc.). The investment costs for each unit were calculated according to the actual capacity, reference size and reference investment cost according to (1):

$$I = I_{old} \cdot \left(\frac{P}{P_{old}}\right)^{n} \cdot F \qquad (1)$$

where I and P denotes the investment cost and size, respectively. The subscripts *old* represent the reference size or cost. F is the overall installation factor, accounting for auxiliary equipment and/or extra costs associated with an investment in that specific unit, and n denotes the scale factor. The investment cost references for the bark boiler and for the major units in the gasification plant is presented in Table 2. The reference investment cost was adjusted to the price level of 2012, by using the ratio between the Chemical Engineering's Plant Cost Index (CEPCI) of year 2012 and the CEPCI of the year in which the reference investments were made.

A cost representing 42% of the equipment and assembly cost was added to cover indirect costs for project administration and development, legal permits, engineering, interest during the construction, insurances, working capital etc. The annual capital cost was estimated by multiplying the annuity factor with the total investment cost (equipment and assembly plus indirect costs). The weighted average cost of capital (WACC) was used as interest rate for the calculation of the annuity factor. The WACC was calculated using the financial parameters specified in Table 3. The annuity factor method was only used to calculate the NH_3 production cost.

The required ammonia selling price to receive a 15% IRR, was calculated using a biomass cost in the range of 11–44 €/MW h (i.e. 50% decrease up to a 100% increase of the reference value). The method for calculating the capital investment cost has an estimated uncertainty in the range of ±30% [31]. The required ammonia selling price to receive 15% IRR, was therefore also investigated for a ±30% change of the gasification plant investment cost.

4. Results and discussion

Two different system configurations using PEBG technology to produce NH_3 were studied, one stand-alone plant (S-A) and one integrated into a pulp and paper mill (Case A). The resulting material and energy flows for the cases are presented in Fig. 3. The overall energy system efficiency (calculated as electricity-equivalents) was estimated using the system boundaries represented by dashed boxes in Fig. 3. The stand-alone plant (S-A) has the same NH_3

Table 2
Investment estimates for the major process units.

Cost reference [Refs.]	Size unit	Ref. size	M€ 2012[a]	Scale factor
Bark boiler [12]	MW_{th}	46	25	0.7
Pre-treatment [27]	Wet t/h	33.5	19	0.8
Air separation unit [27]	t O_2/day	1000	48	0.9
PEBG unit [28]	MW_{th}	0.22	0.5	0.8
Gas cleaning [29]	Dry gas feed kg/s	37	43	0.7
Synthesis plant [30]	TPD NH_3	1800	323	0.7
Off-gas boiler [17]	MW_{th}	355	67	1.0
District heating HEX [17]	MW	355	67	1.0

[a] The reference cost is presented after the conversion to the 2012s price level. Additionally, the overall installation factor (*F*) is included in the reference capital investment.

Table 3
Financial parameters for the value year 2012.

Financial parameter	Value
Bank interest	8%
Equity share of investment	30%
Dept share of investment	70%
Return on equity	15%
Annuity factor	11.8%
Project economic lifetime	20 years
Annual operating hours	8000 h
Annual operating hours, district heating	5000 h

Table 4
Estimates for capital investment costs (M€) and production costs (M€ per year).

	BAU	S-A	Case A
Equipment and assembly	25	300	289
Indirect costs	11	125	121
Total investment cost	36	425	410
Total incremental cost		425	374
Production costs in M€ per year[a]			
Biomass[b]	1.3	43.5	35.6
Power[b]	7.8	19.7	30.2
O&M[c]	1.6	17.8	17.4
Tall oil[d]			−10.0
District heating[e]		−13.1	
Other costs[f]	3.2	0.8	0.8
Subtotal costs (excl. capital)	13.9	68.7	73.9
Annual capital costs	4.2	50.3	48.5
Total production cost	18.1	119.0	122.4
Incremental production cost		119.0	104.3
Production cost (€/t NH_3)		523	458

[a] A negative sign represents revenue.

[b] Biomass cost (22 €/MW h) was taken from [32] and the cost for power (63 €/MW h) was received from [33].

[c] Includes: operations for chemicals, water treatment, insurance, licenses assumed to be 1% of the equipment and assembly. Maintenance, 3% of the equipment and assembly and worker salaries, 6 M€ cost for S-A and Case A.

[d] Tall oil was assumed to be sold as a substitute for heavy fuel oil. The selling cost for tall oil (74 €/MW h) and district heating (70 €/MW h) were taken from [34].

[e] The DH production was calculated to 188 GW h annually for the S-A case.

[f] Ash handling costs was estimated to 57 €/t. In addition, cost for heavy fuel oil in BAU was included.

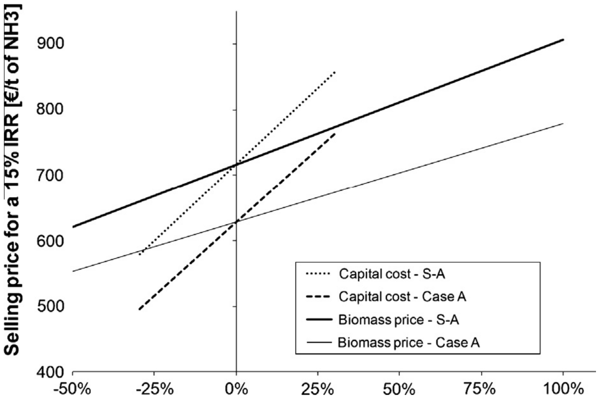

Fig. 4. Required selling price for ammonia for an IRR of 15% when the investment cost and biomass price is varied.

production capacity and annual demand for biomass and electricity as the gasification plant in Case A.

An overall energy system efficiency of 44% (calculated as electricity equivalents) was reached for the non-integrated stand-alone production case (S-A), also accounting for the original operation of the mill (BAU). This result also includes the delivery of nearly 188 GW h of heat per year to the district heating system from the NH_3-plant. In Case A, two additional product streams, NH_3 and tall oil, were added to total mill production. However, one consequence was an increased supply of external biomass to the mill by 50%. Other consequences were decreased electricity production due lower production of HP steam, at the same time as the electricity consumption was almost doubled compared to the reference operation. The calculations showed, however, that the overall system efficiency was increased by 10%-units compared to the reference case. The reasons were mainly that the available internal surplus of biomass (bark) was more effectively used in Case A as well as that tall oil was liberated from the process and became a product output.

The calculated capital investment costs and production costs are presented in Table 4. The gasification train accounts for 18% of the total cost for equipment and assembly. In both cases, a train of four gasifier units were assumed to be required to ensure the high annual operational load (8000 h), where each unit has a capacity of one third of the maximum. A major part (43–44%) of the total investment of equipment and assembly was connected to the NH_3 synthesis unit. The remaining part of the investment cost was more or less equally spread between the fuel pre-treatment unit (12%), the syngas cleaning and conditioning (12%) and the ASU (10–11%).

The economic benefit of integrating the ammonia production compared to the stand-alone case showed to be small as the total production cost (shown in Table 4) was almost equal for both cases. This was mainly due to the significantly higher electricity cost in Case A. The production cost difference of 65 €/t NH_3 can be explained by the avoided costs for the investment in and operation of a new bark boiler.

A NH_3 selling price in the range of 581–882 €/t will be required for the stand-alone plant to receive an IRR between 10% and 20%. Case A consistently required a lower selling price (509–774 €/t of ammonia) for the same IRR. The difference was reduced when a lower return on the invested capital was assumed. The production cost (excluding the cost of capital) for both cases accounted for 34–54% of the ammonia selling price. A selling marginal in the range of 48–66% is therefore required to obtain an IRR of 10–20%. Calculating with the current average selling price of fossil NH_3 (514 €/ton, [26]) an IRR just above 10% was possible to achieve for the integrated system solution, while the stand-alone case reached an IRR of 7%.

Fig. 4 shows the results of the economic sensitivity analysis, where it is illustrated how the biomass price and the investment cost influence, the production cost.

This example, valid for a 15%-IRR, shows that a 30% higher capital cost would increase the required selling price to the same extent as a 95% increase of the biomass price for Case A and a 75% increase for case S-A would. Thus, the capital cost influences the economic profitability in a higher degree than the biomass price.

5. Conclusions

The main objective of this work was to perform a techno-economic evaluation of ammonia production via integrated biomass gasification in an existing pulp and paper mill. The results showed that the overall energy system efficiency was increased by 10%-units (calculated on an electricity equivalence basis) for the integrated system compared to the original operation of the mill in parallel with the operation of a stand-alone ammonia production plant. The economic assessment showed that a higher NH_3 selling price than the current market price was required for the concept to be economically feasible. The integrated case required a selling price in the range of 509–774 €/t to receive an IRR between 10% and 20%. For the stand-alone production plant, an even higher selling price was required. For such IRR-levels, the selling marginal accounted for 48–66% of the NH_3 selling price. Approximately 45% of the total investment cost was directly connected to the investment of the NH_3 synthesis loop, indicating that the production capacities of the studied system configurations (228,000 t/y NH_3) were too small to be viable. Larger production plant capacities would most probably be needed to reach economic feasibility at current NH_3 market price. Lower production costs or higher costs for fossil based alternatives would be required to make biomass-based NH_3 production economically viable.

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